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We introduce and study the concept of transferring the quantum state of two internally-translationally entangled fragments, formed by molecular dissociation, to a photon pair. The transfer is based on intracavity stimulated Raman adiabatic passage.

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The sharing of quantum information (QI) by distant partners is the basis for quantum teleportation [1], cryptography [2] and QI processing [3]. The focus of both theory and experiment has been on the entanglement of *discrete* variables of particles and light [4,5]. An alternative avenue has been opened by the teleportation of *continuous* variables: light-field quadratures [6] or collective atomic spins [7]. It is our aim to explore the *natural* occurrence of inter-particle entanglement in molecular dissociation for QI applications. Entanglement of spin or pseudospin states by dissociation has been studied for some time already [8]. Recently it has been suggested that dissociation into a translationally entangled pair of fragments, followed by a collision of one fragment with an atomic wavepacket, can be used to teleport the wavepacket [9]. Here we put forward and study the concept of transferring the quantum state of two dissociated fragments sharing internal-translational entanglement (ITE) to that of two photons and vice versa.

The following procedure is envisaged for the task at hand (Fig. 1). A cold molecule having velocity  $v_y$  is dissociated, via a single- or two-photon process, to an energy-specific state of two identical molecular or atomic fragments,  $A$  and  $B$ . Each fragment can occupy one of the two internal metastable states, labeled  $|g_1\rangle$  or  $|g_2\rangle$  (even if many internal states are populated by the dissociation process, we can single out the two that satisfy the resonance conditions detailed below). For a given dissociation energy, the fragments' velocities  $\pm v_x$  along the  $x$ -axis depend on the internal excitation state of the system. Therefore we can place two empty optical cavities,  $L$  and  $R$ , aligned along the  $z$  axis, at positions such that only a pair of fragments in their single-excitation state  $|g_1\rangle_A |g_2\rangle_B \pm |g_2\rangle_A |g_1\rangle_B$  enters both cavities, all other outcomes being idle events. This state is entangled and symmetrized or antisymmetrized, depending on the molecular configuration. Under the Raman-resonance condition, each fragment, passing through the sequence of partially overlapping cavity (quantized) and pump (classical) fields, undergoes population transfer to the final state  $|f\rangle$ , via stimulated Raman adiabatic passage (STIRAP) [10], and adds to the corresponding cavity a single photon at a frequency  $\omega_{1,2}^{(L,R)}$  *uniquely determined* by the initial internal and translational state. Upon leaving their cavities through the partially transparent front

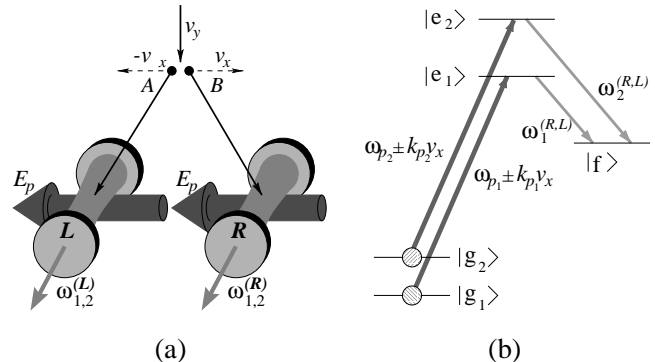


FIG. 1. (a) Dissociating fragments  $A$  and  $B$  pass through the partially overlapping cavities  $L$  and  $R$  and pump fields, respectively, generating two correlated photons. The photons then leak from the cavities through the front mirrors. (b) Level scheme of dissociating fragments.

mirrors, the two entangled photons encode the dissociative state:

$$(|g_1, -p_x\rangle_A |g_2, p_x\rangle_B \pm |g_2, -p_x\rangle_A |g_1, p_x\rangle_B) |0\rangle_L |0\rangle_R \rightarrow |f, -p_x\rangle_A |f, p_x\rangle_B (|\omega_1\rangle_L |\omega_2\rangle_R \pm |\omega_2\rangle_L |\omega_1\rangle_R), \quad (1)$$

where  $\pm p_x$  are the momenta of the fragments in the center-of-mass frame.

Let us now discuss a possible realization of the intracavity Raman resonance conditions [Fig. 1(b)]. Two pump fields, aligned along the  $x$ -axis, with the frequencies  $\omega_{p1}$  and  $\omega_{p2}$  resonantly couple the two ground states  $|g_1\rangle$  and  $|g_2\rangle$  with the excited states  $|e_1\rangle$  and  $|e_2\rangle$ , respectively. Each cavity supports two modes with the frequencies  $\omega_{1,2}^{(L)} = \omega_{p1,2} - k_{p1,2} v_x - (\omega_f - \omega_{g1,2})$  and  $\omega_{1,2}^{(R)} = \omega_{p1,2} + k_{p1,2} v_x - (\omega_f - \omega_{g1,2})$ , where  $\omega_{p1,2} = \omega_{e1,2} - \omega_{g1,2}$ ,  $k_{p1,2} = \omega_{p1,2}/c$  and  $\hbar\omega_j$  ( $j = g_1, g_2, e_1, e_2, f$ ) are the energies of the corresponding atomic states. Thus the mode frequencies of the two cavities are shifted from each other by the difference of the pump-field Doppler shifts for the two fragments,  $\omega_{1,2}^{(R)} - \omega_{1,2}^{(L)} = 2k_{p1,2} v_x$ , while the two modes of each cavity have a frequency difference close to that of the two excited levels,  $\omega_2^{(L,R)} - \omega_1^{(L,R)} = \omega_{e2} - \omega_{e1} \mp (k_{p2} - k_{p1}) v_x \simeq \omega_{e2} - \omega_{e1}$ . This choice ensures the two-photon Raman resonance between either of the states  $|g_1\rangle$  or  $|g_2\rangle$  and the final state  $|f\rangle$  for both fragments.

The Hamiltonian of the system is given by

$$H = \hbar \sum_j \omega_j |j\rangle\langle j| + \hbar \sum_i \{\omega_i a_i^\dagger a_i + [\eta_i(t) |e_i\rangle\langle f| a_i - \Omega_p(t) |e_i\rangle\langle g_i| e^{-i(\omega_{p_i} \mp k_{p_i} v_x)t} + \text{H.c.}]\}, \quad (2)$$

the upper (lower) sign in the exponent standing for fragment  $A$  ( $B$ ) and cavity  $L$  ( $R$ ). Here the first term is the free-fragment Hamiltonian, where the sum is taken over all pertinent states, the second term describes the cavity field,  $a_i$  and  $a_i^\dagger$  ( $i = 1, 2$ ) being the creation and annihilation operators for the corresponding mode, the third term describes the fragment-cavity interaction with the coupling  $\eta_i(t)$  and the last term is responsible for the coupling of the fragment with the classical pump fields having the same Rabi frequency  $\Omega_p(t)$ .

One of the requirements of STIRAP is the “counterintuitive” order of the fields [11], achieved by shifting, by distance  $d$ , the pump field maximum from that of cavity field. For a fragment traveling with the velocity  $v = \sqrt{v_x^2 + v_y^2}$ , the time-dependences of the cavity-(vacuum) and pump fields Rabi frequencies are then given by  $\eta_i(t) = \eta_0^{(i)} \exp[-(vt/w_c)^2]$  and  $\Omega_p(t) = \Omega_0 \exp[-(vt - d)^2/w_p^2]$ , where  $\eta_0^{(i)}$  and  $\Omega_0$  are the corresponding peak Rabi frequencies and  $w_c$  and  $w_p$  are the waists of the cavity and pump fields. During the interaction, the combined system, consisting of the fragment plus its cavity field, will then, under the conditions specified below, adiabatically follow the “dark” eigenstate of the Hamiltonian (2)

$$|u_0^{(i)}(t)\rangle = \frac{\eta_i(t) |g_i, 0\rangle + \Omega_p(t) |f, \omega_i\rangle}{\sqrt{\eta_i^2(t) + \Omega_p^2(t)}}, \quad (3)$$

which does not contain a contribution from the fragment excited state  $|e_i\rangle$ . Thus, the fragment, being initially in state  $|g_i\rangle$ , ends up after the interaction in state  $|f\rangle$ , with a photon added into the corresponding cavity mode  $\omega_i$ .

A standard analysis [11] reveals the following requirements for the system to obey the evolution of the dark state (3): (i) The condition for adiabatic following should be satisfied, namely,  $\eta_0^{(i)} w_c/v, \Omega_0 w_p/v \gg \sqrt{1 + |\Delta_{p_i}| w_{c,p}/v}$ , where  $|\Delta_{p_i}| = k_{p_i} v_x$ . (ii) There has to be sufficient overlap between the two pulses:  $\int \eta_i(t) \Omega_p(t) dt \gg \sqrt{\gamma_{e_i}^2 + \Delta_{p_i}^2}$ , where  $\gamma_{e_i}$  is the decay rate of the excited state  $|e_i\rangle$ . (iii) The fragment-cavity coupling strength should exceed the total relaxation rate of the combined final state  $|f, \omega_i\rangle$ :  $|\eta_0^{(i)}| \gg \gamma_f + 2\kappa$ , where  $\gamma_f$  is the decay rate of the fragment state  $|f\rangle$  and  $\kappa$  is the transmission rate of the cavity field through the mirror. (iv) Finally, the mode spacing of the cavity  $|\omega_2 - \omega_1|$  should exceed both the fragment-cavity coupling strength and the decay rate of the final state:  $|\omega_2 - \omega_1| \gg |\eta_0^{(i)}|, (\gamma_f + 2\kappa)$ . Then, for a given initial

state of the fragment  $|g_i\rangle$ ,  $i = 1$  or  $2$ , the photon will be added only into the resonant mode of the cavity at frequency  $\omega_i$ .

The time-evolution of the system is governed by the master equation  $\partial_t \rho = -i\hbar^{-1}[H, \rho] - \mathcal{L}\rho$ , where  $\mathcal{L}\rho$  describes the fragment and cavity field relaxation processes. In this equation, several additional states of the system, which are decoupled from the Hamiltonian (2), have to be taken into account, as they enter through the possible relaxation channels. These are:  $|f, 0\rangle$ —the fragment is in state  $|f\rangle$  and the cavity is empty;  $|l, \omega_i\rangle$  and  $|l, 0\rangle$ —the fragment is in a low-lying state  $|l\rangle$  to which the state  $|f\rangle$  decays and the cavity has either one or no photon. If the system successfully completes the transfer, then, irrespective of its initial state, it ends up in state  $|l, 0\rangle$  after a time long compared to all relaxation times, which corresponds to the absence of memory in the system about its initial state. The information about the initial state is transferred to the photons emitted by the corresponding cavities, as per Eq. (1).

For numerical simulations we have chosen a well-collimated, cold beam of sodium dimers. Such a beam can be produced via STIRAP photoassociation of cold Na atoms, thereby obtaining translationally cold Na<sub>2</sub> molecules in the chosen vib-rotational state of the electronic ground state  $X^1\Sigma_g^+$  [12]. Subsequently, the inverse STIRAP process dissociates the molecules into pairs of internally-translationally entangled fragments sharing a single excitation [13]. The cavities admit fragments with  $v_x \simeq 5$  m/s and  $v_y \simeq 10$  m/s. The two pump fields couple the two metastable ground states of the Na atom  $|g_1\rangle = |3S_{1/2}, F = 1\rangle$  and  $|g_2\rangle = |3S_{1/2}, F = 2\rangle$  with the excited states  $|e_1\rangle = |4P_{1/2}\rangle$  and  $|e_2\rangle = |4P_{3/2}\rangle$ , respectively. The final state is  $|f\rangle = |4S_{1/2}, F = 2\rangle$ . The sequences of fields seen by the fragments in cavities with parameters similar to those of Ref. [10] are plotted in Figs. 2(a) and 2(b), while the populations of the initial states  $|g_i\rangle$  and the corresponding photon emission rates, defined as  $R_i^{\text{emit}} = 2\kappa(\rho_{f, \omega_i; f, \omega_i} + \rho_{l, \omega_i; l, \omega_i})$ , are plotted in Figs. 2(c) and 2(d). For both cavities, the total photon emission probability  $P_i = \int R_i^{\text{emit}} dt \geq 0.99$ , indicating extremely high efficiency (fidelity) of entanglement transfer between the fragments and the photons, as per Eq. (1). It is noteworthy that this efficiency (or fidelity) remains very high even for considerably lower fragment-cavity coupling strengths (Fig. 2 caption).

The outlined processes are also feasible for molecular dissociation into two molecular fragments in the electronic ground state. As an example consider the photolysis of cyanogen by a 193 nm laser via the reaction channel  $\text{C}_2\text{N}_2(X^1\Sigma_g^+) + \hbar\omega_{\text{diss}} \rightarrow \text{CN}(X^2\Sigma^+, v_1 = 0, N_1 \leq 45) + \text{CN}(X^2\Sigma^+, v_2 = 1, N_2 \leq 31)$ , where  $v_{1,2}$  and  $N_{1,2}$  stand for vibrational and rotational states, respectively, of the electronic ground state  $X^2\Sigma^+$  of CN [14]. By adjusting the positions of the two cavities and the frequencies of the cavity and pump fields, the two ground states

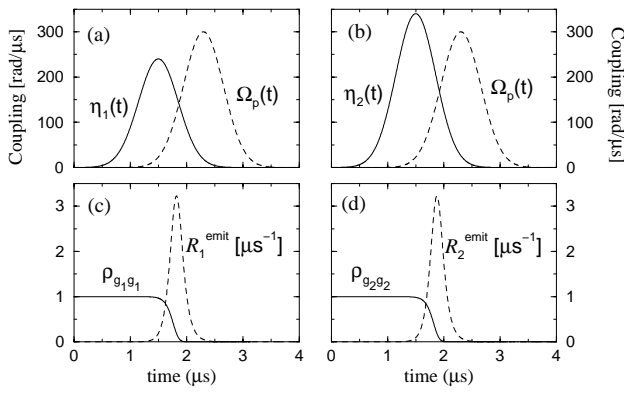


FIG. 2. (a), (b): Time-dependence of the Rabi frequencies of the cavity  $\eta_i(t)$  and pump  $\Omega_p(t)$  fields as seen by fragment  $A$  being initially in state  $|g_1\rangle$  (a) and fragment  $B$  being initially in state  $|g_2\rangle$  (b), or vice versa. (c), (d): Time evolution of the initial-state population  $\rho_{g_1 g_1}$  (c) or  $\rho_{g_2 g_2}$  (d) of the corresponding fragment and emission rate  $R_i^{\text{emit}}$  of the photon from the cavity. The parameters used are:  $\gamma_{e_{1,2}} \simeq 9.6$  MHz and  $\gamma_f \simeq 25$  MHz calculated for the transitions in text,  $\omega_{p_1}/2\pi \simeq 9.0738 \times 10^{14} \text{ s}^{-1}$  and  $\omega_{p_2}/2\pi \simeq 9.0755 \times 10^{14} \text{ s}^{-1}$ ,  $k_{p_{1,2}} v_x/2\pi \sim 15$  MHz,  $(\omega_2 - \omega_1)/2\pi \simeq 168.9$  GHz, corresponding to the mode spacing of a cavity 0.9 mm long. The corresponding coupling constants for the cavity having the mode-waist  $w_c \sim 10 \text{ } \mu\text{m}$  are  $\eta_0^{(1)}/2\pi \simeq 38$  MHz and  $\eta_0^{(2)}/2\pi \simeq 54$  MHz and the cavity linewidth  $2\kappa \simeq 10$  MHz. Reduction of the fragment-cavity coupling constants by a factor of 4 (!) lowers the transfer efficiency from 99% to 90%.

$|g_{1,2}\rangle$  and the final state  $|f\rangle$  can be selected from this vibrational ground state manifold, while the excited states  $|e_{1,2}\rangle$  can be selected from among the excited electronic state manifold  $B^2\Sigma^+$ .

A possible conceptual counterargument for the use of our scheme may be that, depending on the initial state  $|g_1\rangle$  or  $|g_2\rangle$ , the fragment recoil due to the pump-photon absorption will be  $\hbar k_{p_1}$  or  $\hbar k_{p_2}$ , respectively. A subsequent measurement of the fragment's translational state will, in principle, disclose its initial internal state. Consequently, the final motional states of the two fragments will be entangled with the states of the two generated photons, without achieving a complete state mapping from the fragments onto the cavity photons. However, one can easily check that, since the molecule is dissociated in a region having the size  $D_x \leq w_c \simeq 10 \text{ } \mu\text{m}$ , in order to have each dissociating fragment pass through the corresponding cavity waist, the uncertainty of the momentum distribution of the fragment must satisfy  $\Delta p_x \gtrsim \hbar/D_x$ , which is 30 times larger than the photon-recoil difference  $\hbar(k_{p_2} - k_{p_1})$ . Therefore, one will not be able to resolve that difference and deduce the initial state from the fragments' momenta.

We note that a fragment crossing a standing wave cavity at a node, where the electric field amplitude vanishes, will not interact with the cavity mode and the STIRAP

process will not take place. One possibility to overcome this difficulty is to allow the fragment to cross the cavity axis at an angle slightly different from  $90^\circ$ , which can be achieved by tilting the cavity. Another possibility would be to use a running-wave cavity.

Utilizing the transmission protocol of Ref. [4], one may use the generated entangled photon pair to induce the *inverse process at a distant node*, so as to convert the dissociating state  $|f, -p_x\rangle_{A'} |f, p_x\rangle_{B'}$  of another pair of fragments,  $A'$  and  $B'$ , into the initial state of fragments  $A$  and  $B$ . This procedure is applicable to molecular fragments in the electronic ground state, but not to atomic fragments whose final state lifetime  $\gamma_f^{-1}$  is shorter than their time of flight between the dissociation region and the corresponding cavity. Let us therefore consider an alternative *simplified scheme*. Each cavity in Fig. 1(a) now supports only one mode at frequency  $\omega_2^{(L,R)} = \omega_e - \omega_{g_1} \mp k_p v_x$ , respectively. Together with the pump field having a frequency  $\omega_p = \omega_e - \omega_{g_2}$ , this provides the two-photon Raman resonance for each fragment between the states  $|g_2\rangle$  and  $|g_1\rangle$  [Fig. 3(a)]. Upon passing through the cavity and pump fields, only a fragment initially in state  $|g_2\rangle$  will undergo the intracavity STIRAP to state  $|g_1\rangle$  and add a photon to the corresponding cavity. Due to the large two-photon Raman detuning  $\omega_{g_2} - \omega_{g_1} \gg \eta_0, \Omega_0$ , a fragment occupying initially state  $|g_1\rangle$  will exit the interaction region in the *same state*. Thus, both fragments will end up in state  $|g_1\rangle$ . The fragment-cavity Hamiltonian takes the form

$$H = \hbar \sum_j \omega_j |j\rangle\langle j| + \hbar \omega_2 a^\dagger a + \hbar [\eta(t) |e\rangle\langle g_1| a - \Omega_p(t) |e\rangle\langle g_2| e^{-i(\omega_p \mp k_p v_x)t} + \text{H.c.}]. \quad (4)$$

Here we assume that the *momentum uncertainty* of the fragment is *large* and exceeds the photon momentum,  $\Delta p_x > \hbar k_p$ . We, therefore, neglect the recoil of the fragment due to the absorption of the pump photon, obtaining

$$(|g_1, -p_x\rangle_A |g_2, p_x\rangle_B \pm |g_2, -p_x\rangle_A |g_1, p_x\rangle_B) |0\rangle_L |0\rangle_R \rightarrow |g_1, -p_x\rangle_A |g_1, p_x\rangle_B (|0\rangle_L |\omega_2\rangle_R \pm |\omega_2\rangle_L |0\rangle_R). \quad (5)$$

The generated photon leaks out of the corresponding cavity at the rate  $2\kappa$ . Let the output of the cavities  $L$  and  $R$  be directed through, say, an optical fiber into two similar cavities  $L'$  and  $R'$  constituting the receiving node of the system. At that node, a molecule having the same velocity  $v_y$  is dissociated to produce two fragments  $A'$  and  $B'$  in the state  $|g_1\rangle_{A'} |g_1\rangle_{B'}$ . The dissociation energy of the  $A' - B'$  molecule is reduced relative to that of the  $A - B$  molecule, by an amount equal to the energy separation between the two ground states  $|g_1\rangle$  and  $|g_2\rangle$ , so that the dissociating fragments  $A'$  and  $B'$  have the same velocities  $\mp v_x$  as fragments  $A$  and  $B$ . The two pulsed dissociation processes are appropriately synchronized, so

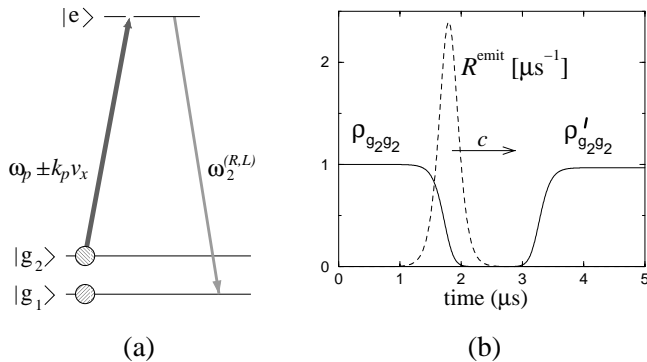


FIG. 3. (a) Level scheme of dissociating fragment interacting with a single mode of the corresponding cavity at frequency  $\omega_2^{(L,R)}$ . (b) Transfer of the population of state  $|g_2\rangle$  between fragments  $A$  ( $B$ ) and  $A'$  ( $B'$ ) via a single photon. The parameters used are:  $\omega_p/2\pi \simeq 5.083 \times 10^{14} \text{ s}^{-1}$ ,  $k_p v_x/2\pi \sim 8.5 \text{ MHz}$ ,  $\eta_0/2\pi \simeq 22 \text{ MHz}$ , and  $\gamma_e \simeq 6.28 \text{ MHz}$ . All other parameters are the same as in Fig. 2.

that the fragments  $A'$  and  $B'$  pass through the cavities  $L'$  and  $R'$  when they receive the output of the cavities  $L$  and  $R$ . Time-reversal is achieved by allowing the fragments  $A'$  and  $B'$  to interact first with the pump field and then with the cavity field. Provided the inversion process is successful, the fragments  $A'$  and  $B'$  will end up in the *same initial internal-translational state* as fragments  $A$  and  $B$  (before the interaction with their cavities).

We have studied the dynamics of the system composed of the two distant nodes using the density operator formalism developed in [15]. The master equation now reads

$$\partial_t \rho = -i\hbar^{-1}[H + H', \rho] - \mathcal{L}\rho - 2\kappa([a'^{\dagger}, \rho] + [\rho a^{\dagger}, a']), \quad (6)$$

where the primed operators stand for the receiving node,  $\mathcal{L}\rho$  describes the fragment and cavity field relaxation processes at both nodes, and the last term provides the unidirectional coupling between the two nodes, in which the output of the cavity at the sending node constitutes the retarded input for the cavity at the receiving node. As expected [4], our simulations show that, provided the photon wavepacket  $R^{\text{emit}}$  is completely time-symmetric, the processes at the two nodes are the time reversals of each other. Figure 3(b) illustrates the results of our calculations. The parameters used again correspond to a dissociating sodium dimer and the excited state  $|e\rangle$  corresponds to the state  $|3P_{1/2}, F=2\rangle$  of the Na atom. The probability of transferring the population of state  $|g_2\rangle$  from fragment  $A$  ( $B$ ) to fragment  $A'$  ( $B'$ ) and thereby achieving the reversal of Eq. (5) is calculated to be 97 %. It is the decay of the excited atomic state  $|e\rangle$  that reduces the fidelity of the process from 100 % to 97 %. Other sources of decoherence, such as photon absorption in the mirror and during propagation, can be accounted for [4] and treated by error correction [3].

To sum up, we have proposed the hitherto unexplored possibility of probing and exploiting the quantum information associated with internal-translational entanglement in molecular dissociation. Our scheme allows, in principle, high-fidelity state transfer from the entangled dissociated fragments to light, thereby producing a highly correlated photon pair. This process, followed by its reversal at a distant node of a quantum network, may have advantageous applications in quantum teleportation, cryptography and communications. Finally, we would like to stress that the proposed scheme requires a combination of processes that have already been experimentally tested [10,11,14].

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